



Time of Emergence of Surface Ocean Carbon Dioxide Trends in the North American Coastal Margins in Support of Ocean Acidification Observing System Design

Daniela Turk^{1,2*}, Hongjie Wang^{3,4}, Xinping Hu⁴, Dwight K. Gledhill⁵, Zhaohui Aleck Wang⁶, Liqing Jiang⁷ and Wei-Jun Cai³

¹ Department of Oceanography, Dalhousie University, Halifax, NS, Canada, ² Lamont-Doherty Earth Observatory, Columbia University, New York, NY, United States, ³ School of Marine Science and Policy, University of Delaware, Newark, DE, United States, ⁴ Department of Physical and Environmental Sciences, Texas A&M University-Corpus Christi, Corpus Christi, TX, United States, ⁵ NOAA Ocean Acidification Program, Silver Spring, MD, United States, ⁶ Department of Marine Chemistry & Geochemistry, Woods Hole Oceanographic Institution, Woods Hole, MA, United States, ⁷ Cooperative Institute for Climate and Satellites, Earth System Science Interdisciplinary Center, University of Maryland, College Park, MD, United States

OPEN ACCESS

Edited by:

Laura Lorenzoni,
University of South Florida,
United States

Reviewed by:

Kim Irene Currie,
National Institute of Water
and Atmospheric Research (NIWA),
New Zealand
Oscar Schofield,
Rutgers, The State University
of New Jersey, United States

*Correspondence:

Daniela Turk
daniela.turk@dal.ca

Specialty section:

This article was submitted to
Ocean Observation,
a section of the journal
Frontiers in Marine Science

Received: 26 October 2018

Accepted: 18 February 2019

Published: 08 March 2019

Citation:

Turk D, Wang H, Hu X,
Gledhill DK, Wang ZA, Jiang L and
Cai W-J (2019) Time of Emergence
of Surface Ocean Carbon Dioxide
Trends in the North American Coastal
Margins in Support of Ocean
Acidification Observing System
Design. *Front. Mar. Sci.* 6:91.
doi: 10.3389/fmars.2019.00091

Time of Emergence (ToE) is the time when a signal emerges from the noise of natural variability. Commonly used in climate science for the detection of anthropogenic forcing, this concept has recently been applied to geochemical variables, to assess the emerging times of anthropogenic ocean acidification (OA), mostly in the open ocean using global climate and Earth System Models. Yet studies of OA variables are scarce within coastal margins, due to limited multidecadal time-series observations of carbon parameters. ToE provides important information for decision making regarding the strategic configuration of observing assets, to ensure they are optimally positioned either for signal detection and/or process elicitation and to identify the most suitable variables in discerning OA-related changes. Herein, we present a short overview of ToE estimates on an OA variable, CO₂ fugacity $f(\text{CO}_{2,\text{sw}})$, in the North American ocean margins, using coastal data from the Surface Ocean CO₂ Atlas (SOCAT) V5. ToE suggests an average theoretical timeframe for an OA signal to emerge, of 23(±13) years, but with considerable spatial variability. Most coastal areas are experiencing additional secular and/or multi-decadal forcing(s) that modifies the OA signal, and such forcing may not be sufficiently resolved by current observations. We provide recommendations, which will help scientists and decision makers design and implement OA monitoring systems in the next decade, to address the objectives of OceanObs19 (<http://www.oceanobs19.net>) in support of the United Nations Decade of Ocean Science for Sustainable Development (2021–2030) (<https://en.unesco.org/ocean-decade>) and the Sustainable Development Goal (SDG) 14.3 (<https://sustainabledevelopment.un.org/sdg14>) target to “Minimize and address the impacts of OA.”

Keywords: ocean acidification, CO₂ fugacity, time of emergence, climate change, novel statistical approaches, observing system optimization, decision making tool

INTRODUCTION

Time of Emergence (ToE) is a term that describes the time when a secular signal emerges from stochastic natural variability. There are two main motivations for determining ToE of ocean observations (Rodgers et al., 2015): to identify when a secular trend unfolding within a marine ecosystem may become evident, relative to the natural background variability, and to inform strategic development of ocean observing systems, purposed for the detection of a secular signal. As a quantitative measure, ToE is a valuable metric to consider asset prioritization in concert with active dialogs with the community of researchers and data users.

Previous studies addressing the concept of emergence [ToE or year of emergence (YoE)] have mostly focused on climate and open ocean variables, such as temperature (Hawkins and Sutton, 2012), precipitation (Mahlstein et al., 2012), and sea levels (Lyu et al., 2014). More recently, ToE has also been investigated for trend signals in ocean biogeochemical variables including key ocean acidification (OA) indicators, mostly for the open ocean using Earth System Models (ESMs) (Ilyina et al., 2009; Friedrich et al., 2012; Christian, 2014; Keller et al., 2014; Rodgers et al., 2015; Carter et al., 2016; Frölicher et al., 2016; McKinley et al., 2016; Henson et al., 2016, 2017; Heinze et al., 2018). These ESM studies have shown that: OA variables experience shorter ToE than most other Essential Ocean Variables (EOVs), such as dissolved organic carbon (DOC) and the biodiversity and ecosystem EOVs (Miloslavich et al., 2018) due to their relatively lower natural variability and stronger non-linear trend in response to anthropogenic forcing; ToE has substantial spatial variability and some studies report relatively shorter ToE in low and high latitudes (Henson et al., 2016, 2017), while others indicate the opposite (Keller et al., 2014); ToE may be more sensitive to stochastic background variability rather than the signal trend strength. Some ocean time-series sites are long enough to detect anthropogenic forcing in ocean biogeochemical variables such as CO₂ fugacity $f(\text{CO}_{2,\text{sw}})$ and pH, apart from natural variabilities (Henson et al., 2016; Bates, 2017). In other systems (e.g., Gulf of Maine Vandemark et al., 2011; Salisbury and Jönsson, 2018), however, detection of OA from atmospheric CO₂ invasion, is complicated by the influence of other factors, such as freshwater input, organic matter cycling, and ocean circulation (Ilyina et al., 2009; Carter et al., 2016).

In the coastal ocean, the estimates of ToE are challenging due to substantially higher variability of OA variables, such as pH (Hofmann et al., 2011; Duarte et al., 2013) and $f(\text{CO}_{2,\text{sw}})$ (Frankignoulle and Borges, 2001; Dai et al., 2009; Guo et al., 2009; Huang et al., 2015), as well as insufficient temporal duration of coastal time-series. The coastal systems are also not suitably resolved in global models to provide reliable ToE estimates. Assuming the anthropogenic forcing is similar in magnitude and direction as in the open ocean, large variability in the coastal ocean should lead to significantly longer ToEs. Kapsenberg and Hofmann (2016) adopted the pH trend value for the North Pacific (Dore et al., 2009; Ishii et al., 2011) and estimated ToE for pH at Anacapa Island (40 years) to be more than triple the time it takes to detect OA trends in the open ocean (Keller et al., 2014). In comparison, for some coastal locations (e.g., Tatoosh Island,

WA, United States), trends may be detected sooner (Wootton and Pfister, 2012). Sutton et al. (2018) used the autonomous moored surface ocean $p\text{CO}_2$ and pH data to derive ToE estimates, suggesting that the time necessary to detect an anthropogenic trend in seawater $p\text{CO}_2$ and pH varies from 8 to 15 years at the open ocean sites, in contrast to coastal sites where estimates ranged from 16 to 41 years.

Several other studies have provided observation-based trend estimates of OA variables for coastal time-series stations or data-driven regional analysis (Wang et al., 2016; Reimer et al., 2017; Wang et al., 2017; Laruelle et al., 2018 and references therein). Using a new statistical approach Generalized Additive Mixed Modeling (GAMM) Wang et al. (2016, 2017) detected multidecadal sea surface $f(\text{CO}_{2,\text{sw}})$ trends at a $1^\circ \times 1^\circ$ resolution. They reported that the sea surface $f(\text{CO}_{2,\text{sw}})$ trends on decadal time scales in the Northern Hemisphere ($1.93 \pm 1.59 \mu\text{atm year}^{-1}$) closely follow the atmospheric $f\text{CO}_2$ increase rate ($1.90 \pm 0.06 \mu\text{atm year}^{-1}$) but appear slower in the Southern Hemisphere ($1.35 \pm 0.55 \mu\text{atm year}^{-1}$). In addition, they also observed differences between western and eastern boundary current-influenced areas. Laruelle et al. (2018) used wintertime data only and investigated the rate of change in air-sea CO₂ gradients in continental shelves and nearby upper slopes. Consistent with previous studies, they found that surface water $p\text{CO}_2$ closely tracks the rate of atmospheric increase in some coastal margins, while other areas have significantly lower or even negative trends as a result of eutrophication (e.g., Baltic Sea) and/or rapid exchange with the open ocean (South Atlantic Bight). Such negative trends may complicate the ToE analysis, as drivers for negative trends [such as gradual changes in precipitation, freshwater input, and net ecosystem production which all could decrease $f(\text{CO}_{2,\text{sw}})$] are challenging to predict on the multidecadal timescale. Therefore, reliably estimating secular trends for the OA variables in coastal regions still remains a great challenge, due to limited spatiotemporal observations relative to the inherent large natural variability of these systems.

Herein, we provide ToE estimates of a variable useful in the detection of OA and related to the inorganic carbon EOV sub-variable $p\text{CO}_2$, $f(\text{CO}_{2,\text{sw}})$, in the North American ocean margins. These estimates are based on both “forced” and “observed” trends, where the latter is calculated using the statistical approach from Wang et al. (2016). The adoption of forced trends allows us to examine whether it is even theoretically possible to detect a long-term $f(\text{CO}_{2,\text{sw}})$ increase due to atmospheric CO₂ invasion amongst natural variability within a suitable timeframe. The comparison between ToEs obtained using the forced and observed trends will help to assess if such trends, based on currently available data, are statistically robust to detect OA or if the signal is muted, masked, or amplified by other forcing(s) that may also be secular and/or oscillating in nature. Regions, identified as departing from the forced OA trends, likely represent areas where discrimination of various contributing forcings needs to be performed before specific attribution of OA can be discerned. The goal of this exercise is to provide strategic information on the most suitable locations for deployment or augmentation of further multidecadal sustained time-series observatories primarily purposed for the detection

of an anthropogenic OA signal (ToE < 20 years). At time-series observatories with longer ToE (> 20 years), the local variability would limit such application without careful discrimination of other processes that affect observed trends. Such locations may, however, represent high priority science sites for other processes. Our approach provides one example of a tool that can be used to aid the prioritization of observation system investments and optimal utilization of resources in the next decade, contributing to the goals of OceanObs19.

DATA AND METHODS

ToE is defined as:

$$\text{ToE} = (N \times \text{noise}) / \text{trend} \quad (1)$$

where noise (μatm) is a measure for natural variability, and N is a specified threshold (weighting coefficient) that characterizes when the “signal” of anthropogenic climate change exceeds the “noise” of natural variability. In past studies, N has been somewhat arbitrarily chosen (as 1, 2, and 3), with 2 used in most studies (Sutton et al., 2018). We also adopt $N = 2$ for this exercise. ToE calculation depends on reliable estimates of both noise and trends. Noise is commonly defined as the standard deviation (SD) of a temporally detrended time-series, which removes the long-term change (temporal trend \times time period). Temporal trends can be calculated over different timescales with a variety of approaches, such as linear least square regression, Markov Chain Monte Carlo (MCMC) method (Majkut et al., 2014), GAMM (see above), and the neural network approach (Landschützer et al., 2013). While temporal trends can be obtained using the actual observational data (Wang et al., 2016, 2017), some studies adopt a “forced” atmospheric CO₂ trend for ToE estimates [e.g., the difference between a constant atmospheric CO₂ at Year 1850 condition and an increasing atmospheric CO₂ (McKinley et al., 2016), or 2 ppm year⁻¹ (Sutton et al., 2018)]. ToE, calculated with the forced trend (ToE_{forced}), represent the timescales for the anthropogenic CO₂ signal, only to become detectable out of internal variability (McKinley et al., 2016) assuming no other secular forcing trends are acting on the system. However, it may be different from the calculated ToE based on observed trends (ToE_{obs}), given that many other processes in addition to atmospheric CO₂ forcing may affect the ocean margins (Fennel et al., 2018).

To better understand the combined effect from both atmospheric CO₂ increase and other forcing, we calculated both ToE_{forced} and ToE_{obs} for $f(\text{CO}_{2,\text{sw}})$ in all $0.5^\circ \times 0.5^\circ$ grids in the North American coastal margins using SOCAT coastal data V5 (Bakker et al., 2016) and the GAMM method. Note that “coastal” is defined as less than 400 km from the coastline, and the data covered the area between 30°S and 70°N. The oceanic CO₂ data collection clearly had monthly bias (with only 85 out of 1341 grids having relatively homogenous monthly samplings). To compensate for the sampling bias, a spline fitting of all de-trended annual data was used to remove seasonal fluctuations in the time-series data in each grid (Wang et al., 2016). The $0.5^\circ \times 0.5^\circ$ grids for ToE calculations were selected based on the following

criteria: (1) have at least a 10-year record; (2) half of the years during the entire time span have data; and (3) have at least six different months of data collection in each grid. For example, a grid that has a 20-year time span will be included in our analysis only if there are at least ten different years and six different months of data coverage. Nevertheless, the majority of grids have more than 10 months of data, which satisfied the requirements. Outliers were defined as any $f(\text{CO}_{2,\text{sw}})$ value falling outside of the 1.5 times interquartile range. These outliers were also excluded from the analysis.

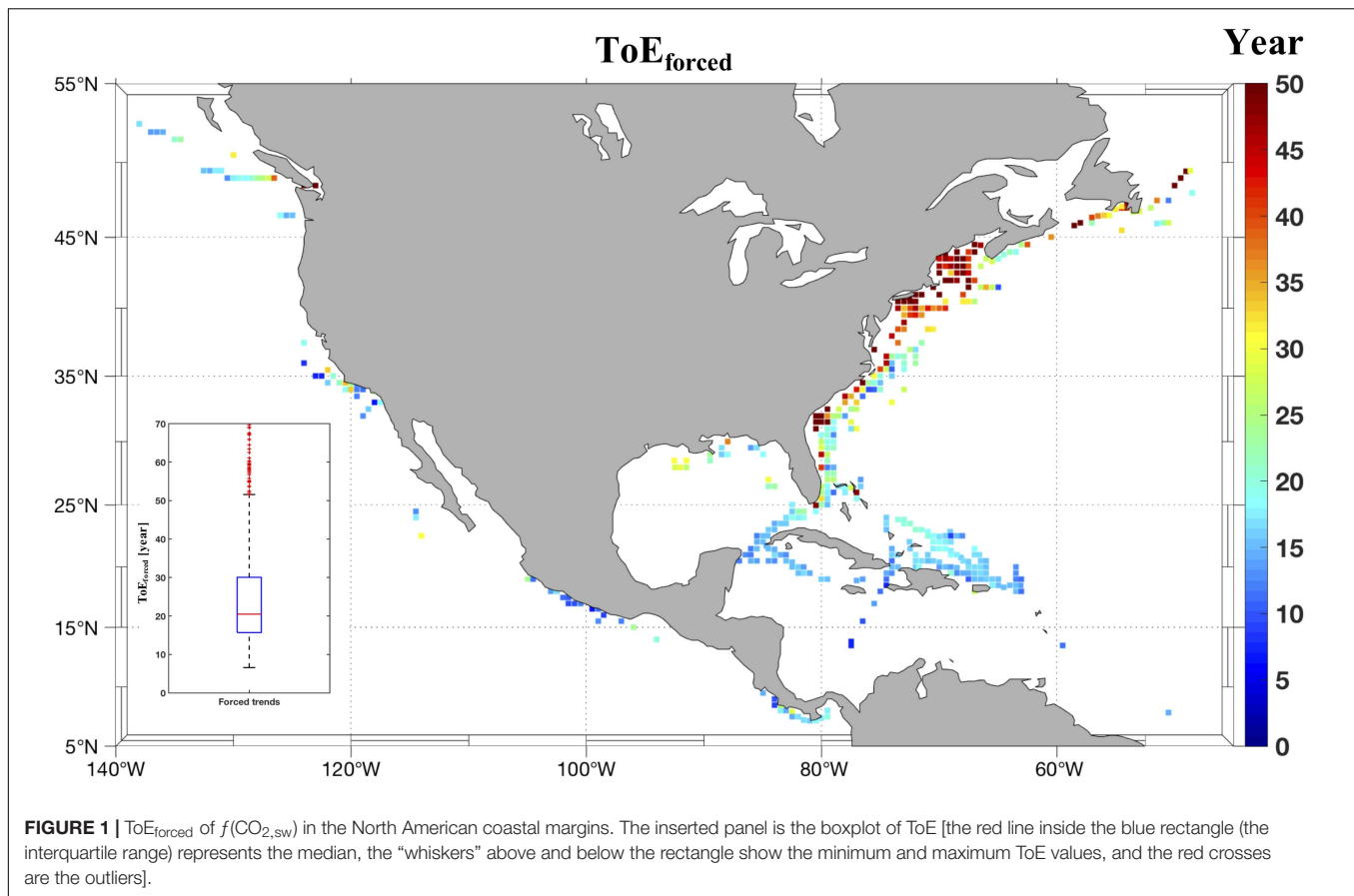
The long-term observed $f(\text{CO}_{2,\text{sw}})$ trend was calculated in each selected grid using the GAMM method as described in Wang et al. (2016). Briefly, the GAMM method predicts $f(\text{CO}_{2,\text{sw}})$ primarily based on three terms derived from observations: seasonal cycle, environmental covariates (temperature and salinity), and the long-term $f(\text{CO}_{2,\text{sw}})$ change. The coefficient of the long-term change represents the $f(\text{CO}_{2,\text{sw}})$ trend during the examined time span. We identified 438 grids with significant “observed” $f(\text{CO}_{2,\text{sw}})$ trends ($p < 0.05$) from 1478 grids in North American coastal margins, and 39 of these 438 grids had negative values. For each of these 438 grids, the long-term change was removed from the original dataset to create the detrended dataset, which was then used to calculate the standard deviation of the detrended time series ($\text{SD}_{\text{detrended}}$). ToE_{obs} was calculated using Eq. 1, as $2 \times \text{SD}_{\text{detrended}} / \text{trend}$. ToE_{forced} was calculated for the same selected grids using Eq. 1 and the forced $f(\text{CO}_{2,\text{sw}})$ trend of 2 $\mu\text{atm year}^{-1}$.

Note that the trends observed may be different from the anthropogenic CO₂ (forced) trend because of the presence or absence of other processes (e.g., upwelling or terrestrial nutrient input). The challenge, however, is to determine whether the observed trend will persist at a constant rate in the future, or if changes in the contribution of the other forcing processes may vary over time.

RESULTS AND DISCUSSION

Spatial Distribution of ToE_{forced} and ToE_{obs}

The average ToE_{obs} of $f(\text{CO}_{2,\text{sw}})$ in the North American coastal margin surface waters (excluding grids with the negative values) is 28.7 ± 20.4 years and significantly ($p < 0.0001$) longer than the average ToE_{forced} 23.0 ± 13.1 years, implying that coastal processes are likely obscuring the signal attributed to the atmospheric CO₂ forcing and making it harder to observe. Both ToE_{forced} (Figure 1) and ToE_{obs} (Figure 2) show high spatial variability but similar spatial patterns with greater values along the east coast (30.7 ± 22.9 years) than in the west coast (23.7 ± 15.4 years), suggesting that observing systems deployed along the Pacific coast will detect the OA signal earlier than other U.S. coastal margins. Relatively larger absolute values of ToE_{obs} in the east coast were partly attributed to higher variability ($\text{SD}_{\text{detrended}}$), as the average observed trends were similar for both coasts ($1.85 \sim 1.88 \mu\text{atm year}^{-1}$). However, other decadal trends may play a role, for example warming, salinity

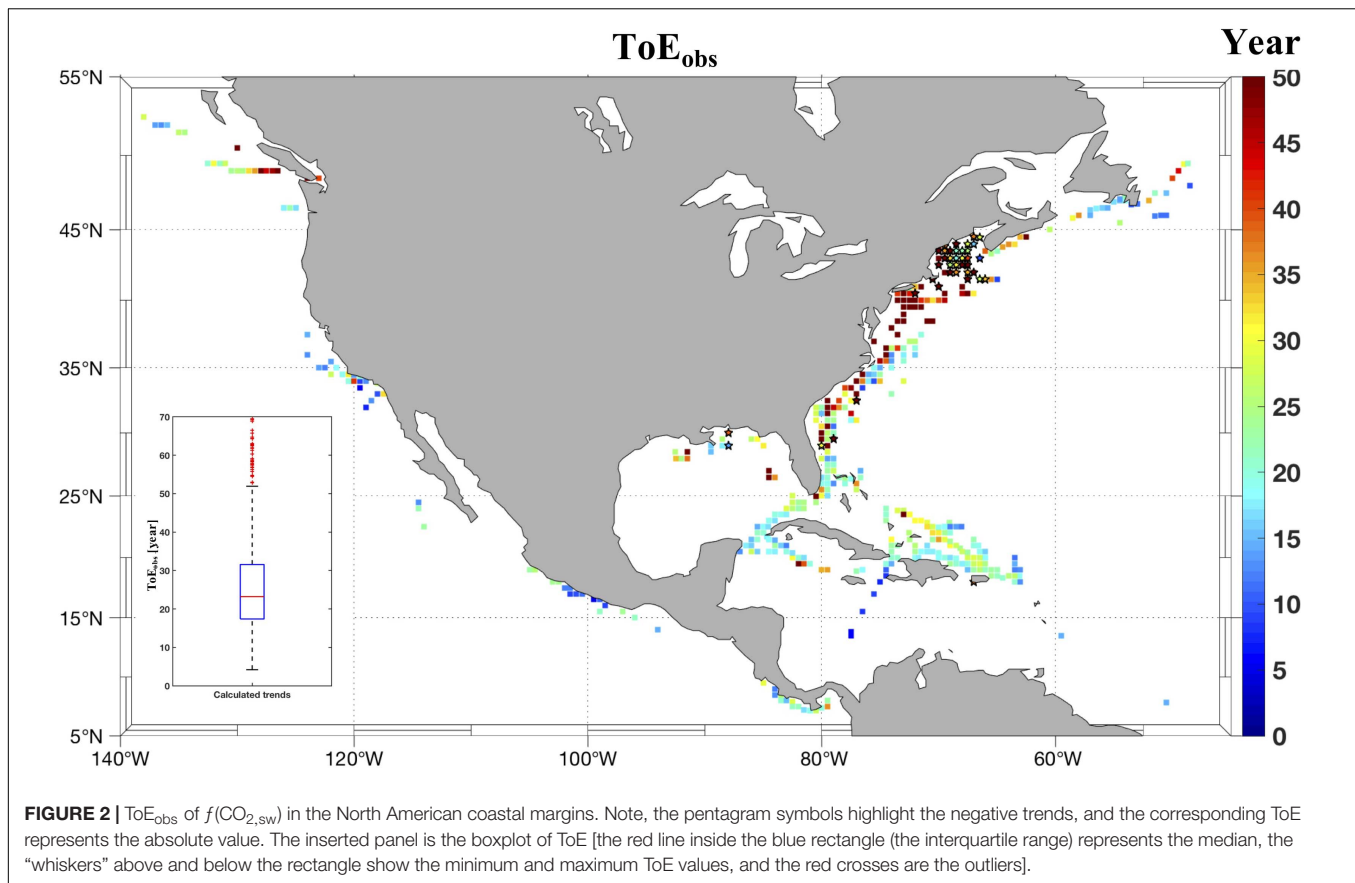


changes and enhanced primary productivity act antagonistically to mask the OA signal in the Gulf of Maine and other northeast regions (Salisbury and Jönsson, 2018), and increased upwelling acts synergistically with OA to amplify the observed trend in the west coast (Fennel et al., 2018 and references therein). The area south of 40°N along the west coast, featured exceptionally short ToE based on both observed and forced trends. For example, the ToE_{obs} in California upwelling areas (17.8 ± 9.1 years) was shorter than other areas, because of low SD_{detrended} ($17.8 \pm 8.8 \mu\text{atm}$). In contrast, in some other areas, such as the Caribbean Sea, the observed trends ($1.5 \pm 0.4 \mu\text{atm year}^{-1}$) were substantially slower than forced trends, which results in a ToE_{obs} almost 6 years longer than would be assumed by OA alone (ToE_{obs} = 21.9 ± 9.6 years versus ToE_{forced} = 16.1 ± 4.0 years). Furthermore, the 39 grids with negative trends are mainly in the Gulf of Maine, along the Gulf Stream and in the northern Gulf of Mexico. In the Northeast U.S. Continental Shelf and Scotian Shelf, such a decrease in surface CO₂ may be due to higher biological uptake as indicated by about 50% increase in surface chlorophyll concentration from 1998 to 2017 (O’Brien, 2018). In the Gulf of Mexico, there were limited data in the current SOCAT database, so a better understanding may be achieved using future iterations that include additional data. It is worth remembering that the calculated ToE_{forced} in these negative CO₂ trends areas only represent a theoretical period, when the anthropogenic CO₂

increases outpace the natural variability, assuming that the drivers are not changing. If the increase of biological uptake of carbon outpaces OA in the Gulf of Maine, as suggested by chlorophyll concentration increasing over the last decade, the OA signal may remain masked.

ToE in the Coastal Margins vs. Open Ocean

In agreement with previous estimates of ToE of OA variables in costal margins (Sutton et al., 2018), our results suggest that the average ToE of surface $f(\text{CO}_{2,\text{sw}})$ in ocean margins is greater (>20 years) than in the open ocean. Ocean margins are subjected to multiple forcings that can confound the $f(\text{CO}_{2,\text{sw}})$ increase caused by atmospheric CO₂ accumulation alone and therefore a longer time-series observation may be needed to detect the OA in comparison to the open ocean, unless efforts are made to deconvolute the various contributing processes. Nevertheless, ToE for $f(\text{CO}_{2,\text{sw}})$ is shorter than that for SST (80.4 ± 49.4 years), consistent with previous studies in the open ocean (Christian, 2014; Keller et al., 2014; Rodgers et al., 2015; Henson et al., 2016, 2017). This is not surprising, considering that CO₂/OA are being directly forced anthropogenically through increasing atmospheric CO₂ concentrations (past centuries), changes in physical oceanographic forcing such as upwelling (recent decades), or nutrient increase (recent decades). SST instead reacts



to the CO₂ increase induced by climate change, and thus the magnitude of the trend signal will be smaller (Keller et al., 2014).

MAIN FINDINGS AND RECOMMENDATIONS

Main Findings

- Based on data currently included in SOCAT V5 coastal database and assuming a forced trend, the theoretical timeframe (ToE_{forced}) within which the OA signal in $f(\text{CO}_{2,\text{sw}})$ will emerge in the North American coastal margins, is 23 ± 13 years on average. This result should be interpreted as an ideal case, whereby the secular trend is solely driven by atmospheric CO₂ invasion amid natural background variability. However, there is considerable fine-scale spatial variability with significantly lower values (<20 years) along the west coast and the Caribbean Sea, while it may take east coast waters 30 years or longer to express OA conditions outside natural variability.
- Less than 30% of the grids provided significant observed $f(\text{CO}_{2,\text{sw}})$ trends, primarily reflecting gaps in the current SOCAT database, or data records of insufficient length, to exhibit the trend. ToE based on these observed trends (ToE_{obs}) was on average 5 years longer than ToE_{forced}, implying that most coastal areas are experiencing

antagonistic secular and/or multi-decadal forcing that dampen the OA signal (notable exceptions are apparent).

- Overall, both ToE_{obs} and ToE_{forced} of surface $f(\text{CO}_{2,\text{sw}})$ in the North American ocean margins (23 ± 13 and 28.7 ± 20.4 years, respectively) are higher than that in the open ocean (<20 years). Large regional differences suggest the influence of long-term non-OA processes (e.g., warming, eutrophication, enhanced primary productivity, and increased upwelling) that may not be sufficiently resolved by current observations.

Recommendations

- ToE_{forced} and ToE_{obs} values suggest that locations along North American west coast will likely express an anthropogenic OA signal sooner, and therefore are suitable places for OA signal detection in the next few decades. At east coast locations with longer ToEs, other forcings may counteract $f(\text{CO}_{2,\text{sw}})$ change. Continuation of time-series observations, which include a comprehensive suite of measurements and/or coupled with targeted process investigations that allow for specific discrimination of the different forcing processes, is needed at these locations to better assess if the observed trends persist and better discern the OA signal.
- In many instances, we cannot expect that the OA signal is currently detectable directly from observing assets. Longer

coastal time-series and the integration of multidisciplinary data from SOCAT, Global Ocean Data Analysis Project (GLODAP), Volunteer Observing Ship (VOS), *in situ* moorings and remote sensing are needed to provide better estimates of ToE_{obs} for $f(\text{CO}_{2,\text{sw}})$ and other OA variables (pH and aragonite saturation states), in order to develop proxies of ToE in areas of limited observations. Closer collaborations between multidisciplinary time-series programs and modelers, who use the observations and coastal modeling that more comprehensively account for the range of coastal biogeochemical processes, are also urgently needed to support decision making tools.

- Further review of methodology for the calculation of trends and variability, to address challenges such as seasonal sampling bias, original vs. detrended data, and use of arbitrary specified threshold (N), is needed. N represents a qualitative assessment of when we suspect the environment experienced by the marine organisms is “noticeably” different. N is most likely species dependent and may vary between different regions, especially if vulnerabilities of organisms are taken into consideration. Studies that will provide better estimates of what a meaningful threshold might be for a specified application are therefore needed. Defining best practices for calculating ToE and standardization of OA measurements is recommended.
- A similar ToE analysis as presented here is recommended for other EOVs in support of designing a new multi-purpose (beyond OA detection) observing system, or augmenting an existing one, to ensure that an optimal set of measurements is included, and the appropriate time frame is planned for.
- Consideration of information such as presented here by the UN Decade of Ocean Science for Sustainable Development (2021–2030) in areas of synthesizing existing research, defining trends, knowledge gaps and priorities for future research, and providing science-based information

to inform managers and policy makers, is recommended. Therefore, there is still a tremendous need for long-term support from funding agencies, as well as initiatives and leadership from both within the oceanographic community and beyond.

DATA AVAILABILITY

Publicly available datasets were analyzed in this study. These data can be found here: <https://www.socat.info>.

AUTHOR CONTRIBUTIONS

DG provided the initial idea for the topic addressed. HW performed the statistical analysis. DT led writing of the manuscript. DT, HW, XH, DG, ZW, LJ, and W-JC contributed to the development, writing, and proofing of this manuscript.

FUNDING

HW was partially supported by an NSF grant (OCE#1654232) while being a research associate at TAMUCC.

ACKNOWLEDGMENTS

We are grateful to Charles Kovach for editorial help and the Ocean Carbon & Biogeochemistry (OCB) Project Office (Heather Benway) for helpful input during the development of an initial idea for this study at the 4th U.S. OA PI meeting sponsored by NSF, NOAA, and NASA. We thank two reviewers and the editor for their insight and comments that improved the manuscript. Lamont-Doherty Earth Observatory contribution number 8290.

REFERENCES

- Bakker, D. C., Pfeil, B., Landa, C. S., Metzl, N., O'Brien, K. M., Olsen, A., et al. (2016). A multi-decade record of high-quality $f\text{CO}_2$ data in version 3 of the surface ocean CO₂ atlas (SOCAT). *Earth Syst. Sci. Data* 8, 383–413. doi: 10.5194/essd-8-383-2016
- Bates, N. R. (2017). Twenty years of marine carbon cycle observations at devils hole bermuda provide insights into seasonal hypoxia, coral reef calcification, and ocean acidification. *Front. Mar. Sci.* 4:36. doi: 10.3389/fmars.2017.00036
- Carter, B. R., Frölicher, T. L., Dunne, J. P., Rodgers, K. B., Slater, R. D., and Sarmiento, J. L. (2016). When can ocean acidification impacts be detected from decadal alkalinity measurements? *Global Biogeochem. Cycles* 30, 595–612. doi: 10.1002/2015GB005308
- Christian, J. R. (2014). Timing of the departure of ocean biogeochemical cycles from the preindustrial state. *PLoS One* 9:e109820. doi: 10.1371/journal.pone.0109820
- Dai, M., Lu, Z., Zhai, W., Chen, B., Cao, Z., Zhou, K., et al. (2009). Diurnal variations of surface seawater $p\text{CO}_2$ in contrasting coastal environments. *Limnol. Oceanogr.* 54, 735–745. doi: 10.4319/lo.2009.54.3.0735
- Dore, J. E., Lukas, R., Sadler, D. W., Church, M. J., and Karl, D. M. (2009). Physical and biogeochemical modulation of ocean acidification in the central North Pacific. *Proc. Natl. Acad. Sci. U.S.A.* 106, 12235–12240. doi: 10.1073/pnas.0906044106
- Duarte, C. M., Hendriks, I. E., Moore, T. S., Olsen, Y. S., Steckbauer, A., Ramajo, L., et al. (2013). Is ocean acidification an open-ocean syndrome? Understanding anthropogenic impacts on seawater pH. *Estuaries Coast.* 36, 221–236.
- Fennel, K., Alin, S., Barbero, L., Evans, W., Bourgeois, T., Cooley, S., et al. (2018). Carbon cycling in the North American coastal ocean: a synthesis. *Biogeosci. Discuss.* doi: 10.5194/bg-2018-420
- Frankignoulle, M., and Borges, A. (2001). The european continental shelf as a significant sink for atmospheric carbon dioxide. *Global Biogeochem. Cycles* 15, 569–576. doi: 10.1029/2000GB001307
- Friedrich, T., Timmermann, A., Abe-Ouchi, A., Bates, N. R., Chikamoto, M. O., and Church, M. J. (2012). Detecting regional anthropogenic trends in ocean acidification against natural variability. *Nat. Clim. Change* 2, 167–171. doi: 10.1038/nclimate1372
- Frölicher, T. L., Rodgers, K., Stock, C., and Cheung, W. W. L. (2016). Sources of uncertainties in 21st century projections of potential ocean ecosystem stressors. *Global Biogeochem. Cycles* 30, 1224–1243.
- Guo, X., Dai, M., Zhai, W., Cai, W.-J., and Chen, B. (2009). CO₂ flux and seasonal variability in a large subtropical estuarine system, the pearl river estuary, china. *J. Geophys. Res.* 114:G03013. doi: 10.1029/2008JG000905

- Hawkins, E., and Sutton, R. (2012). Time of emergence of climate signals. *Geophys. Res. Lett.* 39:L01702. doi: 10.1029/2011GL050087
- Heinze, C., Ilyina, T., and Gehlen, M. (2018). The potential of ²³⁰Th for detection of ocean acidification impacts on pelagic carbonate production. *Biogeosciences* 15, 3521–3539. doi: 10.5194/bg-15-3521-2018
- Henson, S. A., Beaulieu, C., Ilyina, T., John, J. G., Long, M., Séférian, R., et al. (2017). Rapid emergence of climate change in environmental drivers of marine ecosystems. *Nat. Commun.* 8:14682. doi: 10.1038/ncomms14682
- Henson, S. A., Beaulieu, C., and Lampitt, R. (2016). Observing climate change trends in ocean biogeochemistry: when and where. *Global Change Biol.* 22, 1561–1571. doi: 10.1111/gcb.13152
- Hofmann, G. E., Smith, J. E., Johnson, K. S., Send, U., Levin, L. A., Micheli, F., et al. (2011). High-frequency dynamics of ocean pH: a multi-ecosystem comparison. *PLoS One* 6:e28983. doi: 10.1371/journal.pone.0028983
- Huang, W.-J., Cai, W.-J., Wang, Y., Lohrenz, S. E., and Murrell, M. C. (2015). The carbon dioxide system on the mississippi river-dominated continental shelf in the northern gulf of Mexico: 1. Distribution and air-sea CO₂ flux. *J. Geophys. Res. Oceans* 120, 1429–1445. doi: 10.1002/2014JC010498
- Ilyina, T., Zeebe, R. E., Maier-Reimer, E., and Heinze, C. (2009). Early detection of ocean acidification effects on marine calcification. *Global Biogeochem. Cycles* 23:GB1008. doi: 10.1029/2008GB003278
- Ishii, M., Kosugi, N., Sasano, D., Saito, S., Midorikawa, T., and Inoue, H. Y. (2011). Ocean acidification off the south coast of Japan: a result from time series observations of CO₂ parameters from 1994 to 2008. *J. Geophys. Res.* 116:C06022. doi: 10.1029/2010JC006831
- Kapsenberg, L., and Hofmann, G. E. (2016). Ocean pH time-series and drivers of variability along the northern channel islands, California, USA. *Limnol. Oceanogr.* 61, 953–968.
- Keller, K. M., Joos, F., and Raible, C. C. (2014). Time of emergence of trends in ocean biogeochemistry. *Biogeosciences* 11, 3647–3659.
- Landschützer, P., Gruber, N., Bakker, D. C. E., Schuster, U., Nakaoka, S., Payne, M. R., et al. (2013). A neural network-based estimate of the seasonal to inter-annual variability of the atlantic ocean carbon sink. *Biogeosciences* 10, 7793–7815.
- Laruelle, G. G., Cai, W.-J., Hu, X., Gruber, N., Mackenzie, F. T., and Regnier, P. (2018). Continental shelves as a variable but increasing global sink for atmospheric carbon dioxide. *Nat. Commun.* 9:454. doi: 10.1038/s41467-017-02738-z
- Lyu, K., Zhang, X., Church, J. A., Slangen, A. B. A., and Hu, J. (2014). Time of emergence for regional sea-level change. *Nat. Clim. Change* 4, 1006–1010. doi: 10.1038/nclimate2397
- Mahlstein, I., Hegerl, G., and Solomon, S. (2012). Emerging local warming signals in observational data. *Geophys. Res. Lett.* 39, L21711.
- Majkut, J. D., Sarmiento, J. L., and Rodgers, K. B. (2014). A growing oceanic carbon uptake: results from an inversion study of surface pCO₂ data. *Global Biogeochem. Cycles* 28, 335–351.
- McKinley, G. A., Pilcher, D. J., Fay, A. R., Lindsay, K., Long, M. C., and Lovenduski, N. S. (2016). Timescales for detection of trends in the ocean carbon sink. *Nature* 530, 469–472. doi: 10.1038/nature16958
- Milosavlach, P., Bax, N. J., Simmons, S. E., Klein, E., Appeltans, W., Aburto-Oropeza, O., et al. (2018). Essential ocean variables for global sustained observations of biodiversity and ecosystem changes. *Global Change Biol.* 24, 2416–2433. doi: 10.1111/gcb.14108
- O'Brien, T. (2018). *Indicator: Ocean Chlorophyll Concentrations*. Available at <https://data.globalchange.gov/report/indicator-ocean-chlorophyll-concentrations-2018>
- Reimer, J. J., Wang, H., Vargas, R., and Cai, W.-J. (2017). Multidecadal fCO₂ increase along the united states southeast coastal margin. *J. Geophys. Res. Oceans* 122, 10061–10072.
- Rodgers, K., Lin, J., and Frolicher, T. L. (2015). Emergence of multiple ocean ecosystem drivers in a large ensemble suite with an Earth system model. *Biogeosciences* 12, 3301–3320.
- Salisbury, J. E., and Jönsson, B. F. (2018). Rapid warming and salinity changes in the gulf of maine alter surface ocean carbonate parameters and hide ocean acidification. *Biogeochemistry* 141, 401–418. doi: 10.1007/s10533-018-0505-3
- Sutton, A. J., Feely, R. A., Maenner-Jones, S., Musielwicz, S., Osborne, J., Dietrich, C., et al. (2018). Autonomous seawater pCO₂ and pH time series from 40 surface buoys and the emergence of anthropogenic trends. *Earth Syst. Sci. Data Discuss.* doi: 10.5194/essd-2018-114
- Vandemark, D., Salisbury, J. E., Hunt, C. W., Shellito, S. M., Irish, J. D., McGillis, W. R., et al. (2011). Temporal and spatial dynamics of CO₂ air-sea flux in the gulf of maine. *J. Geophys. Res.* 116:C01012. doi: 10.1029/2010JC006408
- Wang, H., Hu, X., Cai, W.-J., and Sterba-Boatwright, B. (2017). Decadal fCO₂ trends in global ocean margins and adjacent boundary current-influenced areas. *Geophys. Res. Lett.* 44, 8962–8970. doi: 10.1002/2017GL074724
- Wang, H., Hu, X., and Sterba-Boatwright, B. (2016). A new statistical approach for interpreting oceanic fCO₂ data. *Mar. Chem.* 183, 41–49. doi: 10.1016/j.marchem.2016.05.007
- Wootton, J. T., and Pfister, C. A. (2012). Carbon system measurements and potential climatic drivers at a site of rapidly declining ocean pH. *PLoS One* 7:e53396. doi: 10.1371/journal.pone.0053396

Conflict of Interest Statement: The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

Copyright © 2019 Turk, Wang, Hu, Gledhill, Wang, Jiang and Cai. This is an open-access article distributed under the terms of the Creative Commons Attribution License (CC BY). The use, distribution or reproduction in other forums is permitted, provided the original author(s) and the copyright owner(s) are credited and that the original publication in this journal is cited, in accordance with accepted academic practice. No use, distribution or reproduction is permitted which does not comply with these terms.